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# Reactions of Cyclic Oxalyl Compounds XXXIX [1]. Reactions of 4-Ethoxycarbonyl-5-phenyl-2,3-dihydrofuran-2,3-dione with Heterocumulenes and *Schiff* Bases

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**Summary.** Furan-2,3-dione **1** reacts with arylisocyanates to the corresponding pyrrol-2,3-diones **2**, whereas conversion with diisopropylcarbodiimide affords the oxazepin-6,7-dione derivative **3** in 68% yield. 1,3-Oxazines **5**, **6**, and **7** were obtained by thermolysis of **1** in boiling xylene in presence of arylisocyanates, diphenylketen-*p*-tolylimine, and *Schiff* bases, most likely by trapping the α-oxoketene intermediate **4**. Preparative flash vakuum pyrolysis (FVP) of **1** and **2b** gave **8** and **9**, respectively.

**Keywords.** Furan-2,3-dione; Pyrrol-2,3-diones; 1,3-Oxazepine; Thermolysis; Cycloaddition.

**Reaktionen cyclischer Oxalylverbindungen, 39. Mitt.** [1]. **Umsetzungen von 4-Ethoxycarbonyl-5-phenylfuran-2,3-dion mit Heterocumulenen und *Schiff*schen Basen**

**Zusammenfassung.** Das Furan-2,3-dion **1** reagiert mit Arylisocyanaten zu den entsprechenden Pyrrol-2,3-dionen, wohingegen mit Diisopropylcarbodiimid das 1,3-Oxazepinderivat **3** in 68%iger Ausbeute gebildet wird. Die 1,3-Oxanzine **5**, **6** und **7** werden durch Thermolyse von **1** in siedendem Xylol in Gegenwart von Arylisocyanaten, Diphenylketen-*p*-tolylimin und *Schiff*schen Basen erhalten, offensichtlich durch Abfangen des intermediär gebildeten α-Oxoketens **4**. Präparative Flash-Vakuum-Pyrolyse (FVP) von **1** bzw. **2b** ergaben **8** bzw. **9**.

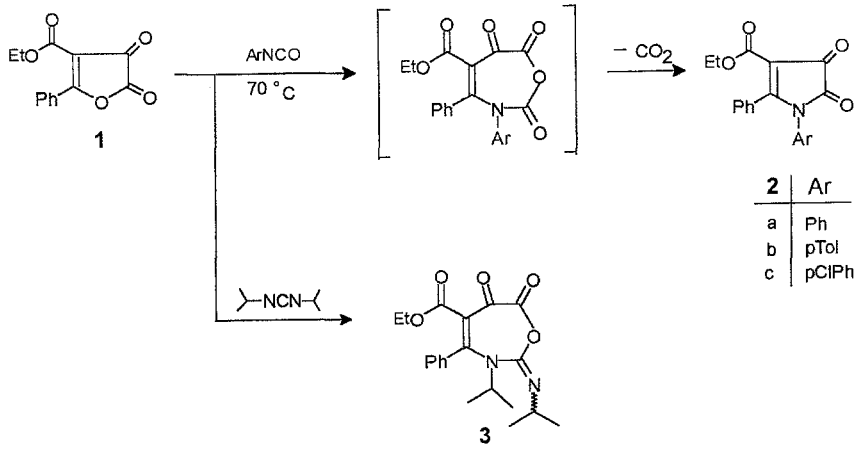
## Introduction

Furan-2,3-diones in general are considered as convenient and versatile synthons in heterocyclic synthesis [2]. Depending on the substitution pattern at C-4, they can serve as hetero-diene systems in various cycloaddition processes, usually accompanied by surprising rearrangements [3]. Furthermore, these molecules are suitable precursors in generating highly reactive α-oxoketenes during simple thermolysis in solution [4] or, employing the flash vacuum pyrolysis methodology [5], in some cases leading to remarkably stable representatives of this class of compounds [6]. α-Oxoketenes can be stabilized both sterically and electronically [6]; in particular, ketene carboxylic acid derivatives are extraordinarily stable [7]. Therefore, in order to extend our investigations on such furan-2,3-diones suitable

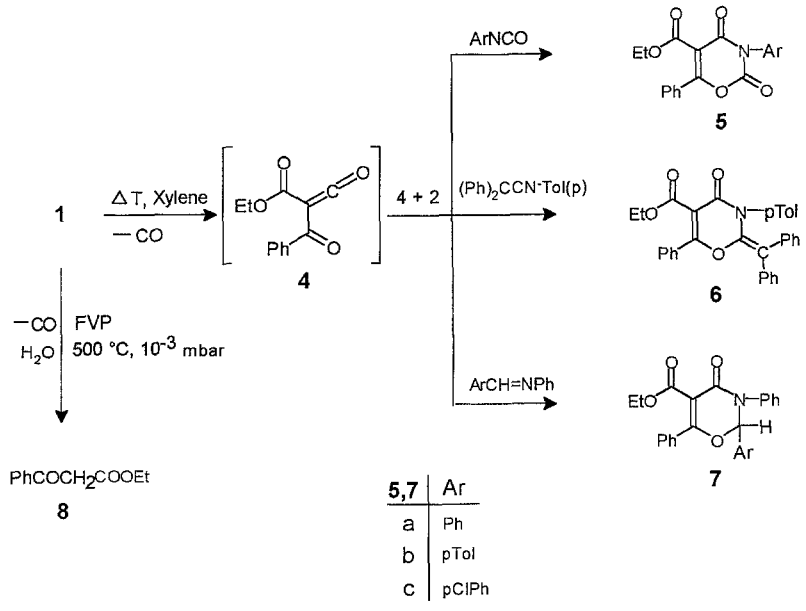
as potential precursors to generate stable $\alpha$-oxoketenes, we have examined 4-ethoxycarbonyl-5-phenyl-furan-2,3-dione (first prepared by *Saalfrank et al.* [8]), not only with respect to its pyrolytic properties but also regarding its behaviour in cycloaddition reactions with some polar double bond systems, *e.g.* heterocumulenes and *Schiff* bases.

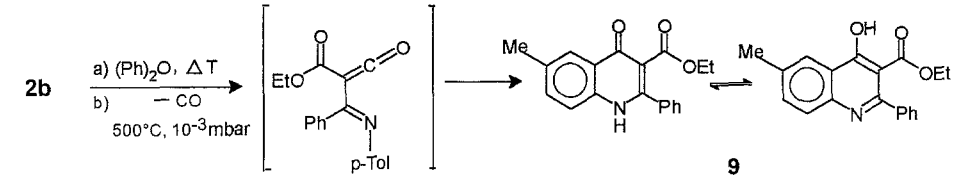
## Results and Discussions

Treatment of the yellow furandione **1** with arylisocyanates at 70°C furnishes the corresponding red coloured pyrrol-2,3-diones **2**. Their structural confirmation was derived mainly from comparison of their analytical and IR spectroscopic data with those of an authentic sample synthesized in a different cyclocondensation reaction of the corresponding enaminoester and oxalylchloride [9] as well as with further compounds containing the pyrrol-2,3-dione moiety [10]. Surprisingly, employing diisopropylcarbodiimide in reaction with **1**, the 1,3-oxazepin derivative **3** is obtained exclusively. The formation of this 1:1 adduct is strongly supported by the results of all analytical and spectroscopic measurements, in particular by the presence of three carbonyl bands (IR: 1715, 1690, and 1660 $cm^{-1}$; $^{13}C$ NMR: 190.8, 163.8, 161.5 ppm) and the absence of any quarternary $sp^3$ carbons attached to heteroatoms as well as any NH or OH groups. It is interesting to note that from the reaction of 4-benzoyl-5-phenylfuran-2,3-dione and phenylisocyanate the corresponding pyrrol-2,3-dione is obtained as a side product only (15%), whereas the main product (40%) is a pyrrolopyrimidine formed *via* [4 + 2] cycloaddition of the heterocumulene to the oxa-1,3-diene moiety of the educt, accompanied by a novel furandione rearrangement [3e, 11]. Furthermore, products obtained from reactions of 4-benzoyl substituted furan- or pyrroldiones with diisopropylcarbodiimide implicate a similar reaction pathway. Obviously, as expected, the ester carbonyl group in **1** is not sufficiently active to serve as part of the heterodiene unit group for hetero-*Diels-Alder* reactions. As a consequence, an alternative reaction pathway leading to **2** and **3**, respectively, should imply insertion of the heterocumulene into the furan ring, thus forming a seven-membered ring system which in case of isocyanates decarboxylates affording the pyrroldiones **2**. In case of the carbodiimide, the product is stable (**3**).



The reaction of furandione **1** with several heterocumulenes as well as with *Schiff* bases under thermolytic conditions in solution (boiling xylene) afforded the corresponding 1,3-oxazine derivatives **5–7** in moderate to acceptable yields (35–70%), obviously formed *via* a [4 + 2] trapping process of $\alpha$-oxoketene **4** as highly reactive intermediate [12, 13].




The presence of the 1,3-oxazin-2,4-dione moiety in **5** is confirmed by its characteristic IR and $^{13}C$ spectroscopic data (see also Ref. [13]): three carbonyl absorption bands (1790–1680 $cm^{-1}$) as well as the corresponding signals in the $^{13}C$ NMR spectra (**5a**: $\delta$ = 162.8, 162.1, 159.3 ppm; **5b**: $\delta$ = 164.8, 163.9, 161.2 ppm). Applying diphenylketen-*p*-tolylimine as dienophile leads to isomers depending on addition to the C=N or C=C double bond, respectively. However, the 1,3-oxazine unit in **6** was verified unambiguously from $^{13}C$ NMR data compared with those of a very close analogue ($\delta$ in brackets) obtained from a similar reaction employing 4-benzoyl-5-phenylfurandione instead of **1** [14]: C-2: 158.9 (159.5), C-4: 164.3 (163.0), C-5: 117.6 (118.1), C-6: 143.1 (142.9), exocyclic $C(Ph)_2$: 107.0 (110.0) ppm. In particular, the absence of any $sp^3$ carbon atom excludes addition to the C=C double bond as well as any possible *Dimroth* rearranged products [15, 16]. It should also be pointed out that only few [4 + 2] cycloaddition reactions of ketenimines involving their C=N-double bond have been reported so far [6, 16]. Furthermore, addition of ketenimines to 1,3-oxadienes proceeding as discussed above is also favoured by semiempirical molecular orbital calculations (AM1, [17]).

2,3-Dihydro-1,3-oxazin-4-ones **7** were obtained from addition of *Schiff* bases to **1** in 55 – 68% yield. From analytical and spectroscopic data (see Experimental), in

particular from the $^1$H NMR spectrum of **7**, important structural information can be obtained. $\delta$ values of 6.45–6.55 ppm for the protons at C-2 are in good accordance with those of very close analogues having identical 2*H*-1,3-oxazin-4-one ring systems ([13]: 6.64, 6.71 ppm; [18]: 6.9 ppm).

Finally, flash vacuum pyrolysis (FVP) experiments using furan-2,3-dione **1** as well as pyrrol-2,3-dione **2b** (pyrolysis of **2a** has already been reported [19]) as potential precursors to generate the corresponding neat $\alpha$-oxoketene or imidoylketene, respectively, failed. Whereas from pyrolysis of **1** benzoylethylacetate **8** could be isolated, obviously obtained as a result of hydrolysis of the primary formed $\alpha$-oxoketene during work-up from the cold finger, in case of **2b** the imidoylketene generated immediately undergoes a $6\pi$-electrocyclic ring closure to afford the quinoline derivative **9** [20] during warm-up. This behaviour is well known from other N-aryl-imidoylketenes [19, 21], irrespective from which precursor they were generated and independent of the thermolytic or pyrolytic reaction conditions employed. A clear distinction between the tautomeric forms of **9** from simple IR and NMR spectroscopic data [22] is difficult. However, exchange of the acidic hydrogen against deuterium and its influence on the chemical shift values as well as signal intensities in the $^{13}$C NMR spectrum clearly indicate that the 4-quinolone tautomer obviously predominates. In particular, the signals at 149.5 (m, C-2, $\Delta\delta = 0.2$ ppm) and 139.6 (t, C-8a, $\Delta\delta = 0.2$ ppm), exhibit a significant decrease of intensity when deuterated due to loss of relaxation pathways [23].

## Experimental

Melting points were obtained on a Gallenkamp melting point apparatus Mod. MFB-595 (open capillary tubes). IR spectra (cm$^{-1}$) were measured on a Perkin-Elmer Model 298 IR spectrometer (KBr pellets), $^1$H and $^{13}$C NMR spectra were recorded on a Varian Gemini 200 instrument (*TMS* as internal standard, $\delta$ in ppm). Microanalyses were performed on a C,H,N-Automate Carlo Erba 1106.

*1-Aryl-4-ethoxycarbonyl-5-phenyl-1H-pyrrol-2,3-diones* (**2**)

*General procedure:* Add 1 mmol of furandione **1** [8] to 1 mmol of the corresponding isocyanate and keep the reaction mixture for 2 h at 70°C until the evolution of gas stops. After addition of 5 ml of dry toluene, the solid products **2** precipitate as red crystals in 75–85% yield.

*4-Ethoxycarbonyl-1,5-diphenyl-1H-pyrrol-2,3-dione* (**2a**) [9]

Yield: 0.258 g (80%); red prisms; mp.: 173°C (toluene); Ref. [9]: m.p.: 191°C.

*4-Ethoxycarbonyl-5-phenyl-1-(4-methylphenyl)-1H-pyrrol-2,3-dione* (**2b**)

Yield: 0.285 g (85%); red prisms; mp.: 189–190°C (toluene); IR: 1780 ms, 1720s (C=O); $C_{20}H_{17}NO_4$ (335.36); calcd.: C 71.64, H 5.11, N 4.18; found: C 71.82, H 5.07, N 4.11.

*4-Ethoxycarbonyl-5-phenyl-1-(4-chlorophenyl)-1H-pyrrol-2,3-dione* (**2c**)

Yield: 0.266 g (75%); red prisms; mp.: 186–188°C (toluene); IR: 1770 ms, 1715s (C=O); $C_{19}H_{14}NO_4Cl$ (355.78); calcd.: C 64.13, H 3.96, N 3.93, Cl 9.97; found: C 63.98; H 3.94, N 3.90, Cl 10.21.

*5-Ethoxycarbonyl-3-isopropyl-2-isopropylimino-4-phenyl-1,3-oxazepine-6,7-dione* (**3**)

1 mmol of furandione 1 is dissolved in 1 mmol of N,N-diisopropylcarbodiimide, and the reaction mixture is kept at room temperature for 12 h. Then, ether/petrolether (40–60°C) 1:1 is added slowly until the product **3** precipitates which is recrystallized from ether/petrolether after suction.

Yield: 0.253 g (68%); white powder; mp.: 135°C; IR: 2980 m, 1715s, 1685s, 1660s; $^1$H NMR ($CDCl_3$): $\delta = 1.26$ (dt, 9H, $3CH_3$), 1.39 (d, 6H, $2CH_3$), 4.10–4.45 (m, 4H, 2CH, $CH_2$), 7.40–8.10 (m, 5H, arom); $^{13}$C NMR: $\delta = 190.6$ (C=O), 163.7, 161.5 (OC=O), 148.1 (C-2), 142.4 (C-4), 138.5, 136.5, 131.9, 130.7 (Ph), 117.0 (C-5), 63.8 ($OCH_2$), 49.5, 48.2 (CH), 26.1, 21.0, 15.9 ($CH_3$); $C_{20}H_{24}N_2O_5$ (372.39); calcd.: C 64.44 H 6.49 N 7.51; found: C 64.60, H 6.67, N 7.39.

*1,3-Oxazines* (**5–7**)

*General procedure:* 1 mmol of furandione **1** is dissolved in 15 ml of boiling xylene. Then, a solution of 1 mmol of the corresponding heterocumulene or *Schiff* base, respectively, dissolved in 3 ml of xylene, is added dropwise through 15 min. Refluxing is continued for 1.5 h, then the solvent is removed *in vacuo*, and the residual oil is treated with ether/petrolether (40–60°C) to give the crude products which are recrystallized from suitable solvents. In case of the ketenimine, the crude residue is separated and purified by dry column flash chromatography (eluant: toluene/chloroform 10:1, $R_f = 0.3$).

*5-Ethoxycarbonyl-3,6-diphenyl-4H-1,3-oxazine-2,4-dione* (**5a**)

Yield: 0.23g (67%); colourless solid; m.p.: 164°C (ethanol); IR (KBr): 1785s, 1740s, 1680s, 1640; $^1$H NMR ($CDCl_3$): $\delta = 1.22$ (t, $CH_3$), 4.28 (q, $CH_2$), 7.34–7.74 (m, 10H, arom); $^{13}$C NMR ($CDCl_3$): $\delta = 162.8$, 162.1 (C-4, ester-CO, exchangeable), 159.2 (C-6) 147.2 (C-2), 133.3 (N-Ph), 109.3 (C-5); $C_{19}H_{15}NO_5$ (337.33); calcd.: C 67.64, H 4.84, N 4.15; found: C 67.87, H 4.48, N 4.11.

*5-Ethoxycarbonyl-3-(4-methylphenyl)-6-phenyl-4H-1,3-oxazine-2,4-dione* (**5b**)

Yield: 0.246 g (70%); white solid; m.p.: 190°C (ethanol); IR (KBr): 1775s, 1720s, 1690s, 1640s; $^1$H NMR ($CDCl_3$): $\delta = 1.24$(t, $CH_3$), 2.41(s, $CH_3$), 4.31 (q, $CH_2$), 7.22–7.73 (m, 9H, arom); $^{13}$C NMR ($CDCl_3$): $\delta = 162.7$, 161.8 (C-4, ester-CO), 158.1 (C-6), 147.3 (C-2), 139.5 (N-Ph), 130.6, 129.0 (Ph, 4-MePh) 109.25 (C-5); $C_{20}H_{17}NO_5$ (351.34); calcd.: C 68.36, H 4.78, N 3.98; found: C 68.27, H 4.95, N 3.80.

*5-Ethoxycarbonyl-3-(4-chlorophenyl)-6-phenyl-4H-1,3-oxazine-2,4-dione* (**5c**)

Yield: 0.242 g (65%); white solid; m.p.: 200°C (ethanol); IR (KBr): 1790m, 1735s, 1690s, 1650s; $^1$H NMR ($CDCl_3$) $\delta = 1.22$ (t, $CH_3$), 4.30 (q, $CH_2$), 7.22–7.73 (m, 9H, arom); $^{13}$C NMR ($CDCl_3$): $\delta = 164.2$, 164.1 (C-4, ester-CO), 160.9 (C-6), 148.5 (C-2), 137.6, 134.6, 133.6 (qu. aryl-C), 110.7 (C-5); $C_{19}H_{14}ClNO_5$ (371.78); calcd.: C 61.38, H 3.79, N 3.76; found: C 61.09, H 3.73, N 3.66.

*5-Ethoxycarbonyl-3-(4-methylphenyl)-6-phenyl-2-diphenylmethylene-4H-1,3-oxazine-2,4-dione* (**6**)

Yield: 176 g (35%); yellow crystals; m.p.: 152°C (toluene); IR (KBr): 1730s, 1685s, 1650s; $^{13}$C NMR ($CDCl_3$); $\delta = 165.6$, 164.3 (C-4, ester-CO), 158.9 (C-6), 143.09 (C-2), 137.7, 137.0, 135.8, 135.7, 132.3, 130.5 (Ph-C)′ 117.57 (C-5), 107.03 (*exo*-C); $C_{33}H_{27}NO_4$ (501.54); calcd.: C 79.02, H 5.42, N 2.79; found: C 79.22, H 5.42, N 2.75.

*5-Ethoxycarbonyl-2,3,6-triphenyl-2H,1,3-oxazine-4-one* (**7a**)

Yield: 0.232 g (58%); white solid; m.p.: 173°C (ethanol); IR (KBr): 1745s, 1685s, 1655s; $^1$H NMR ($CDCl_3$): $\delta = 1.18$ (t, 3H, $CH_3$), 4.11 (q, 2H, $CH_2$), 6.45 (s, 1H, H-2 oxazine), 7.34–7.74 (m, 15H, arom); $C_{25}H_{21}NO_4$ (399.41); calcd.: C 75.17, H 5.29, N 3.30; found: C 74.96, H 5.18′ N 3.46.

*5-Ethoxycarbonyl-2-(4-methylphenyl)-3,6-diphenyl-1,3-oxazine-4-one* (**7b**)

Yield: 0.261 g (63%); white solid; m.p.: 187°C (ethanol); IR (KBr): 1725s, 1680s, 1650s; $^1$H NMR ($CDCl_3$): $\delta = 1.18$ (t, 3H, $CH_3$), 2.31 (s, 3H, $CH_3$), 4.11 (q, 2H, $CH_2$), 6.55 (s, 1H, H-2 oxazine), 7.13–8.01 (m, 14H, arom); $C_{26}H_{23}NO_4$ (413.44); calcd.: C 75.52, H 5.60, N 3.38; found: C 75.63, H 5.69, N 3.35.

*5-Ethoxycarbonyl-2-(4-chlorophenyl)-3,6-diphenyl-1,3-oxazine-4-one* (**7c**)

Yield: 0.239 g (55%); white solid; mp.: 198°C (EtOH); IR (KBr): 1735s, 1680s, 1650s; $^1$H NMR ($CDCl_3$): $\delta = 1.18$ (t, 3H, $CH_3$), 2.31 (s, 3H, $CH_3$), 4.11 (q, 2H, $CH_2$), 6.55 (s, 1H, H-2 oxazine), 7.13–8.15 (m, 14H, arom); $C_{25}H_{20}ClNO_4$ (433.91); calcd.: C 69.19, H 4.64, N 3.22, Cl 8.18; found: C 68.96, H 4.55, N 3.16, Cl 7.95.

*3-Ethoxycarbonyl-6-methyl-2-phenyl-4(1H)-quinolin-4-one* (**9**)

Refluxing 1 mmol of **2b** in 20 ml of diphenyl ether for 25 min and subsequent cooling of the reaction mixture to room temperature, followed by addition of 20 ml *n*-hexane, afforded the crude product as a white solid which was collected by filtration and washed with ethanol to give 0.29 g (95%) of pure **9**.

Mp.: 259°C (ethanol; Ref. [20]: m.p.: 253–254°C); IR (KBr): 3300–3050 (OH), 1720 (OC=O), 1625, 1550 (C=O) $cm^{-1}$[22]; $^1$H NMR (*DMSO*-$d_6$): $\delta = 0.84$ (t, 3H, $CH_3$), 2.38 (s, 3H, $CH_3$), 3.91 (q, 2H, $CH_2$), 7.18–7.90 (m, 3H, arom); $^{13}$C NMR (*DMSO*-$d_6$): $\delta = 173.5$ (d, C-4), 166.1 (t, ester-CO), 149.31 (m, C-2), 139.4 (t, C-8a), 115.5 (s, C-3); $C_{19}H_{17}NO_3$ (307.33); calcd.: C 74.25, H 5.57, N 4.55; found: C 74.77, H 5.69, N 4.54.

*Flash vacuum pyrolysis of* **1**

0.246 g (1 mmol) of furan-2,3-dione **1** were pyrolyzed at 500°C/$10^{-3}$ mbar for 5 h. The product (**8**; 0.125 g, 65%) was isolated from the cold finger after warm-up, washing with ether, and evaporation of the solvent. **8** was identified by comparison with an authentic sample.

*Flash vacuum pyrolysis of* **2b**

Employing the same procedure as described above, 0.22 g (72%) of **9** were isolated from 0.335 g (1 mmol) of **2b** and identified by comparison with an authentic sample.

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